# Extremely BRIGHT Incredibly *FAST*

*Multidisciplinary teams of researchers are leading cutting-edge experiments at world-class, light-source facilities to observe how materials change under extreme conditions.*

WHAT began as a ten-centimeter contraption made of wire, glass, and red sealing wax that used electrical and magnetic fields to accelerate protons in a spiral-shaped path before they collided with a target—the first cyclotron—spawned larger, more powerful devices for groundbreaking discoveries; unlocked the secrets of the atom; and revealed new elements, isotopes, cosmic particles, and antiparticles. Upon receiving the 1939 Nobel Prize in Physics for the invention and development of the cyclotron, Ernest O. Lawrence anticipated that his "proton merry-go-round" was still in its nascent stage. He explained, "We have been looking towards the new frontier in the atom, the domain of energies above a hundred-million volts, for we have every reason to believe that there lies ahead for exploration a territory with treasures transcending anything thus far unearthed. To penetrate this new frontier will require the building of a giant cyclotron, perhaps weighing more than 4,000 tons…We have been working on the designs of such a great instrument and are convinced that there are no insurmountable technical difficulties in the way of producing atomic projectiles of energies well above one hundred million volts."

More than 80 years later, some of the largest, most complex machines on the planet—synchrotron radiation facilities that generate the world's brightest x rays—stand as a legacy of Lawrence's work and the result of progressive scientific discoveries yielding more powerful cyclotrons, particle accelerators, and x rays. These synchrotron radiation light sources produce "hard" x rays at the high-energy, short-wavelength band of the electromagnetic spectrum as well lower energy "soft" x rays. At wavelengths of 0.10 to 0.01 nanometers, comparable to interatomic distances, hard x rays are ideal for studying atoms; soft x rays measure about 1 nanometer in length and are perfect for studying biological samples, nano structures, and energy science in general. Their brilliance—which varies depending on the beamline and the facility—allows them to penetrate matter and interact with atoms, producing x-ray diffraction, imaging, and spectroscopy patterns that researchers examine to understand how elements react under extreme conditions. Meeting the Laboratory's missions requires that researchers understand how materials behave under high pressure or forces, specifically, how their atomic structure changes, how quickly, if kinetics is involved in those changes, and what new material properties emerge. "The truth is, how a big, giant bomb behaves depends directly on the behavior of infinitesimal atoms," says Livermore materials scientist Mukul Kumar. Laboratory researchers work at the atomic scale on time-resolved, light-source experiments to observe how materials transform under extreme conditions. Determining when a substance will shift between states of solid, liquid, gas, or even plasma in some cases plays a significant role in the materials and modeling simulation codes the Laboratory uses to verify the safety, security, and effectiveness of the nation's nuclear weapons stockpile in the absence of testing. The goal of these experiments is to capture the data needed to validate the physics models in the simulation codes, and Livermore's scientists interrogate and resolve the discrepancies. "The predictions are only as good as the underlying science; the physics must be right. These light-source experiments are the proving ground, and the Laboratory's modeling and simulation capabilities provide the connective tissue that informs our large-scale hydrodynamics testing, our detonation science, and materials deformation studies."

"No matter the specifics of the experiment, we're always pushing these materials into extreme conditions to see how they change on timescales from microseconds to nanoseconds–100 billionths of a second. To observe what's happening, we need x rays that have short pulse lengths, appropriate energies, and sufficient intensities to capture and document those changes just as fast," says Trevor Willey, physicist in the Materials Science Division at the Laboratory. Some light-source experiments involve shock compression using a gas gun, some involve a laser or laser-shock compression in combination with a diamond anvil cell (DAC), and some experiments involve detonations. "Each of these light sources, beamlines, and even experimental hutches along the beamlines produces experiments with specific flavors," says Willey.

## The Right Flux

"One of the biggest challenges with these experiments is that atoms react in nanoseconds. The light-source facility needs to produce x rays at that same speed. The flux of photons—the number of photons per second in a given area—needs to rise above the background noise of the system in order for us to document that reaction. The flux of photons must be good enough to capture the behavior of the atoms," says Kumar. The flux at the Advanced Photon Source (APS) at Argonne National Laboratory (ANL) outside of Chicago, Illinois, is well-suited for a variety of experiments designed by Livermore researchers and scientists.

Ultrabright, high-energy, storage-ring-generated x-ray beams arrive at "sectors" distributed across APS. One of those sectors is the High-Pressure Collaborative Access Team (HPCAT) at Sector 16 where four simultaneously operational beamlines have been established with an array of x-ray probes and diagnostics optimized for high-pressure research using x-ray diffraction, x-ray spectroscopy, and x-ray imaging techniques. Lawrence Livermore, Los Alamos, and Sandia national laboratories and the Stewardship Science Academic Alliances Program are principal stakeholders in the HPCAT team; the National Nuclear Security Administration (NNSA) is the primary sponsor. "Before the COVID-19 pandemic, HPCAT hosted about 750 on-site experimental users annually, and 50 percent of these experimenters were academics, students, and postdoctoral researchers from around the world, many of whom wind up working for the national labs," says Nenad Velisavljevic, director of HPCAT and a Livermore employee. "Over the last two years, HPCAT implemented

The first successful cyclotron (above) built by Ernest O. Lawrence (below) accelerated a few hydrogen ions to an energy of 80,000 electronvolts. The device earned Lawrence $500 from the National Research Council towards the construction of a machine that might be useful for nuclear physics. (Images courtesy of University of California, Lawrence Berkeley National Laboratory.)

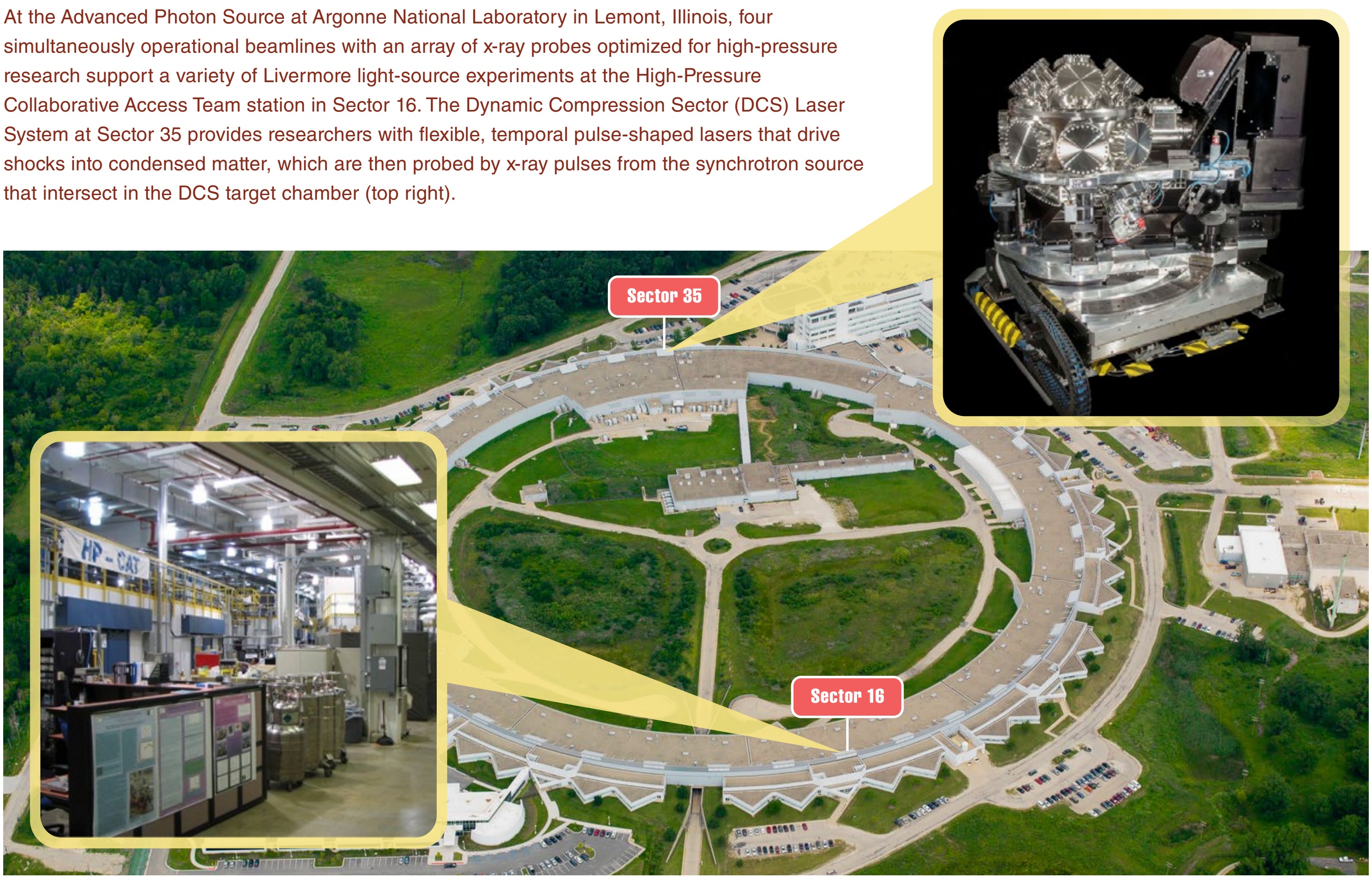
critical and necessary adjustments and now hosts remote-user operations as well. HPCAT continues to provide critical capabilities for NNSA laboratories even with the ongoing COVID restrictions."

Livermore physicist Samantha Clarke has utilized HPCAT's high flux to enable more accurate safety and performance modeling of nitroamine CL-20, a powerful explosive important to weapons research, by probing CL-20's structural properties under extreme conditions. Clarke and her team determined that the compressibility of the molecule along each axis is consistent across the entire pressure range, potentially due to the molecule's cagelike structure. Based on the diffraction data captured, which matches calculated values, the team identified novel experimental equation-of-state parameters describing how CL-20 changes under different conditions. "The experiments at HPCAT allow us to analyze high-pressure and temperature states of matter that no one in the world has ever investigated with such high resolution. For many of these experiments, we are observing never-before-seen phases," says Clarke.

## Research Hutches

APS's Dynamic Compression Sector (DCS) consists of a series of hutches at Sector 35 where Livermore scientists perform a variety of powder gun, gas gun, and laser-driven research using permanently installed drivers. The user-defined DCS B Station has offered Livermore scientists the flexibility to explore aspects of high-explosive detonation physics over the last seven years. Conventional high explosives usually have some excess carbon, and under the extreme temperatures and pressures of detonation, a Livermore team has observed the formation of carbon nanoparticles in diamond, graphitic, and even liquid carbon phases depending on the pressures and temperatures attained. In one case, analysis of time-resolved, small-angle, x-ray scattering data during detonation of a hydrogen-free explosive, dinitrofurazanfuroxan, yielded dynamic measurements of liquid-carbon condensation and solidification into nano-onions—concentric series of spherical carbon shells roughly 30 nanometers in diameter—in just over 200 nanoseconds. (See *S&TR,* July 2017, pp. 12–15.)

At the Advanced Photon Source at Argonne National Laboratory in Lemont, Illinois, four simultaneously operational beamlines with an array of x-ray probes optimized for high-pressure research support a variety of Livermore light-source experiments at the High-Pressure Collaborative Access Team station in Sector 16. The Dynamic Compression Sector (DCS) Laser System at Sector 35 provides researchers with flexible, temporal pulse-shaped lasers that drive shocks into condensed matter, which are then probed by x-ray pulses from the synchrotron source that intersect in the DCS target chamber (top right).



Observing the type of carbon produced and how quickly it forms over hundred-nanosecond timescales provides both empirical input and validation of detonation codes essential to stockpile stewardship. "Using small angle scattering, we can observe the emergence and aggregation of these particles during detonation," says Willey. "One of the most fascinating and technologically promising class of particles we're looking at is nano diamonds. One of the most common methods used to produce nano diamonds is through high-explosive detonations, where the temperature and pressure is intense enough to form diamond. Contrary to what's out there in the scientific literature, these nano diamonds aggregate during detonation nearly as fast as they form in this hot carbon soup. Understanding how these detonations produce nano diamonds may inform how we tweak their properties." These nano diamonds have several technological applications. Nano diamonds can absorb substantial heat, efficiently emit electrons, and are also inert and nontoxic. Rigid-yet-tunable nano diamonds come in a variety of shapes and sizes and have the potential to revolutionize pharmaceuticals, fuel additives, material coatings, other nanotechnologies, and even quantum computing. (See *S&TR,* March/April 2008, pp. 14–16.)

The DCS C-Hutch at APS houses high-energy lasers that Livermore scientists use to shock-compress materials. Dynamic compression experiments occur on nanosecond timescales and require a light source that can generate extremely bright short-pulsed x rays so scientists can take a "snapshot" of the sample in its extreme state. For one such experiment, Livermore physicist Richard Briggs proposed shock-compressing gold, which had no expected phase transitions but the test would provide a strong signal from the scattered x rays. To the team's surprise, the sample's crystal structure changed at close to 200 gigapascals (GPa)—almost two-thirds the pressure at the center of the Earth—the first time this phase had ever been observed in gold. The team's findings led to additional experiments designed to investigate high-pressure phase transitions in materials regularly used at Lawrence Livermore's National Ignition Facility and at the University of Rochester's Omega Laser Facility in Rochester, New York. "At DCS, we've laid the groundwork for performing these experiments and developed unique techniques that apply the Laboratory's mission for understanding materials response relevant to stockpile stewardship. And we're still finding new surprises!" says Briggs.

Livermore scientists (left to right) Federica Coppari, Amalia Fernandez Panella, Amy Coleman, and Samantha Clarke at the Dynamic Compression Sector of the Advanced Photon Source.

## Nanosecond Laser Compression

"What we really want to know about material behavior is how quickly the products form and what happens to them," says high-energy-density physicist Jon Eggert. Light sources like the Linac Coherent Light Source (LCLS) in Menlo Park, California, an Office of Science User Facility operated for the U.S. Department of Energy by Stanford University, allow Livermore scientists to directly measure material behavior under laser-driven dynamic compression and answer these questions in unprecedented detail. "LCLS produces a different flavor of x rays—shorter, more intense, more monochromatic, more coherent, but with a slower repetition rate," says Willey. The LCLS delivers 120 x-ray pulses per second, each lasting one quadrillionth of a second (femtoseconds), the rate at which atomic movements can be tracked and measured. These measurements help constrain material behavior models used by the programs in several Laboratory applications including weapons design, advanced materials and manufacturing, and computer modeling and simulations.

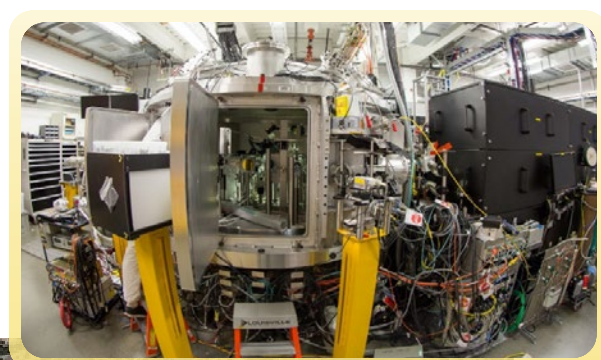
At the Matter in Extreme Conditions End Station at LCLS, Livermore physicists Martin Gorman, Samantha Clarke, Jon Eggert, and Ray Smith have teamed up with other experimentalists from around the world to subject zirconium samples to laser-driven shock compression to 22 GPa so they transition to the high-pressure *omega* phase that forms in zirconium alloys with most transition metals. For each experiment, a laser beam 500-micrometers in diameter drove a shock wave into zirconium samples using a 10-nanosecond flat-top laser pulse. The extreme pressures produced by the laser-driven compression shock exceed pressures produced by static compression methods or dynamic compression using gas guns, generating novel structures. "By using one of the fastest, brightest x-ray sources in the world, we can collect atomic snapshots of samples after they have been compressed by a laser-induced shockwave. The x-ray snapshots indicate if the sample has transformed to a new phase at high pressure, which may have vastly different material properties to the ambient material," says Gorman. "This opens the door to recovering novel phases of matter with functional properties. Crucially, nanosecond laser compression allows access to vast regions of phase spaces that cannot be studied using other recovery techniques."

Livermore physicist Amy Coleman led a team that developed a new way of housing potassium in a target package for light source experiments so they could study potassium under dynamic compression at LCLS without it reacting, either with the air or the other target components. Very reactive and difficult to study using static compression techniques, potassium can eat away at the diamonds in DACs causing experimental failure. Potassium also melts at a relatively low temperature, so much of the collected diffraction data describes its liquid state. Analysis of liquid diffraction is complex and requires diffraction data of the highest quality. "At LCLS we are able to probe the liquid structure of potassium at pressures and temperatures that had never been accessed. The breadth of the detectors, the high brilliance, and the short duration of the x rays allow us to obtain data of high-enough quality to perform quantitative analysis on liquids via dynamic compression. We are paving the way to obtaining structural information and measurements of density for a wide range of materials in the liquid state and broadening understanding of myriad extreme condition liquid systems such as planetary interiors," says Coleman.

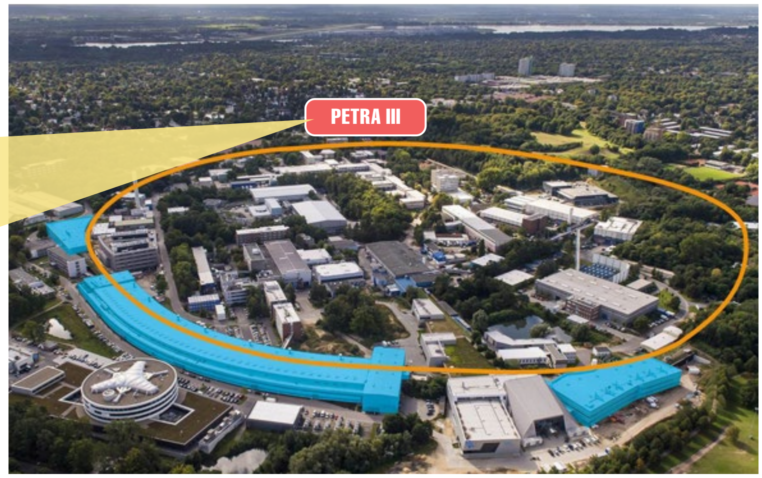
## Transformative Tools

In 2007, Livermore physicist Will Evans and his team pioneered the invention of a device for studying the dynamic-pressure properties of materials—the dynamic diamond anvil cell (dDAC)—that repetitively applies time-dependent load and pressure to a sample by adapting electromechanical piezoelectric actuators to a conventional diamond anvil cell. The dDAC allows the study of phase transition kinetics and metastable phases at strain rates of up to 500 GPa per second through precise specification. Initially, the dDAC was used with laboratory-based optical spectroscopy diagnostics and optical imaging techniques because extant x-ray light sources and detectors had slow frame rates, which produced poor time resolution. Then, in 2019, Livermore physicist Zsolt Jenei and his team in the Lawrence Livermore High-Pressure Physics Group, along with scientists from Deutsches Elektronen-Synchrotron (DESY), the European Synchrotron Radiation Facility, and the universities of Oxford, Bayreuth, and Goethe developed a next-generation dDAC that can compress samples faster than any other previous DAC—1.6 billion atmospheres per second. (See *S&TR*, July/August 2019, pp. 20–23.)

The improved dDAC characterizes the response of a sample under well-controlled fast compression. "Our technique can dial in different compression rates from slow to fast, which cannot be done with laser compression or a gas gun," says Jenei. "This allows us to bridge the gap between traditional static and shock compression and investigate compression-rate-dependent phenomena across orders of magnitude, but we also need an x-ray source with enough photons to capture the diffraction and sensitive enough to detect changes at almost the exact same speed." Since the invention of the dDAC, one challenge has been conducting fast-diffraction experiments due to the lack of photon flux—the number of photons per second, per unit area—and fast, highly sensitive, high-energy x-ray diffraction detectors.



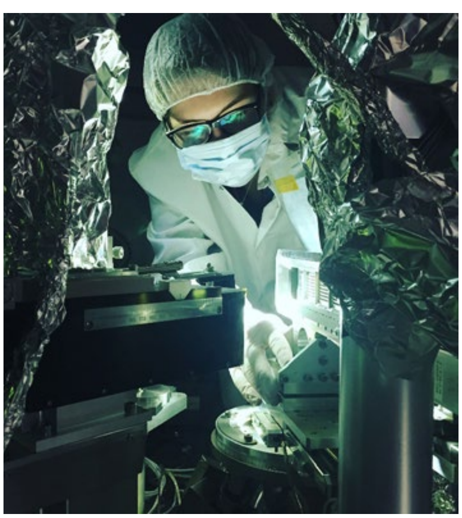

The Matter in Extreme Conditions experimental hutch (right) at SLAC's Linac Coherent Light Source (LCLS) in Menlo Park, California (below), allows Livermore scientists to directly measure material behavior under laser-driven dynamic compression using 120 x-ray pulses that last one quadrillionth of a second, the rate at which atomic movements can be tracked and measured.




Using a next-generation dynamic diamond anvil cell, the LAMBDA detector (left), and the Extreme Conditions Beamline at PETRA III (right), the Livermore team has achieved unprecedented compression rates of up to 160 terapascals per second on a sample of gold.

With the arrival of a high-brilliance third-generation synchrotron radiation source at DESY's PETRA III, outside of Hamburg, Germany, the biggest and most brilliant light source in the world, and the development of the gallium-arsenide (GaAs) LAMBDA detector, Livermore scientists can collect diffraction images with adequate short exposure times and temporal resolution. Using next-generation dDAC and the Extreme Conditions Beamline, the team has achieved compression rates of up to 160 terapascals per second on a sample of gold. More recently, the team explored the effect of kinetics related to nucleation and growth of different high-pressure phases of bismuth at various compression rates. They used time-resolved x-ray diffraction with first-time 0.25 millisecond time resolution to accurately determine phase-transition pressures at compression rates spanning five orders of magnitude while compressing a sample of bismuth. "In the relatively low pressure and temperature region, bismuth has a complex phase diagram. Under dynamic compression from the ambient state, the incommensurate phase III of bismuth has not been observed. We were surprised to see that an overpressurization of bismuth's third and fourth phase boundary happens at fast compression rates for different bismuth samples and stress states," says Jenei.

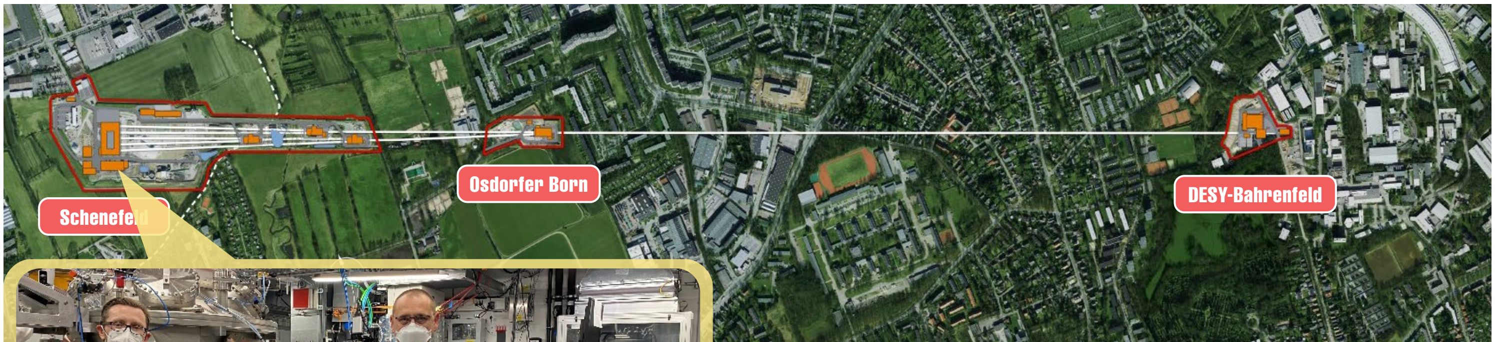
These surprising findings also uncover new avenues for future studies of transition kinetics at previously inaccessible compression rates. One of the challenges of these experiments is combining techniques so that they work simultaneously with the dDAC. "On their own, each technique is relatively straightforward, but setting everything up to start at the exact same moment can be challenging," says Earl O'Bannon, a Livermore staff scientist in the High-Pressure Physics Group who prepares the samples and runs the experiments on the DESY PETRA III Extreme Conditions Beamline. "It's exciting to be constantly pushing the edge of what's possible with the diamond anvil cell. We are now able to use the x rays to obtain structural information and directly image opaque metal samples using x-ray imaging techniques while achieving millisecond time resolution. We also get to see ideas go from the drawing board to reliable tools and techniques that support the Laboratory's stockpile stewardship mission, find widespread use in the high-pressure community, and maintain Livermore's leading role in high-pressure science," says O'Bannon.



Livermore physicist Amy Coleman loads a target mount into the vacuum chamber at the LCLS. The experiments are typically conducted under vacuum to avoid unwanted effects from the presence of air during laser ablation of the targets. Researchers typically load several cartridges at a time so that they can shoot around 200 targets before the chamber is vented and new targets are put in place.

## Unexpected Changes

Using a unique combination of a short-pulse optical laser and an ultrashort, free-electron laser pulse at the Pohang Accelerator Laboratory X-ray Free Electron Laser in South Korea, Livermore physicist Hyunchae Cynn and his international colleagues recorded the atomic structural evolution of shock-compressed iron at the remarkable time resolution of 50 picoseconds under a high-strain rate. The team documented a three-wave temporal evolution of the elastic, plastic, and deformational phase transitions to the second phase, followed by post-compression phases. Their experiment was the first direct, complete




Livermore scientists (left, from left to right) Earl O'Bannon, Zsolt Jenei, and Daniel T. Sneed stand in front of the high-energy-density (HED) instrument at the European X-ray Free Electron Laser (Eu-XFEL) facility in Schenefeld, Germany (above). The team performed the first dynamic diamond anvil cell experiments on the HED instrument at the Eu-XFEL supporting a team of more than 50 international scientists.

The Advanced Light Source at Lawrence Berkeley National Laboratory in Berkeley, California. (Photo credit: University of California, Lawrence Berkeley National Laboratory.)

Lawrence Livermore materials scientist, Holly Carlton (above), prepares a sample at the Advanced Light Source at the Lawrence Berkeley National Laboratory beamline 8.3.2.


The Pohang Accelerator Laboratory X-ray Free-Electron (PAL-XFEL) Laser (above) in Pohang, South Korea outside of Seoul provides XFEL radiation in a range of 0.1 to 6 nanometers. (Photo credit: POSTECH.)

observation of shock wave propagation associated with crystal structural changes captured by high-quality x-ray diffraction data. The experiment also demonstrated the ability to measure atomistic evolution during the lattice compression and release processes at unprecedented time and strain rate. “Pohang is an exceptional facility that combines shock compression of materials using an optical laser at the picosecond range that is able to accurately capture and measure high-resolution data. We really didn’t expect to see so many changes in such a short time,” says Cynn.

Cynn joined Livermore colleagues, Jenei, O’Bannon, Evans, and Magnus Lipp to explore the structural evolution of tantalum at high pressure and temperature by irradiation using the x-ray free electron laser (XFEL) Beam at European XFEL (Eu-XFEL) in Schenefeld, Germany. Their approach used the x rays to directly heat tantalum under extreme pressures in a DAC. The applied XFEL beam also provided x-ray diffraction information and density of tantalum as a function of pressures and temperatures. The XFEL’s brilliance allowed the team to measure a dynamic material response within a 10-femtosecond x-ray pulse. Due to the ultrashort x-ray pulse structure at Eu-XFEL, material change—lattice expansion, melting, and chemical reaction—induced by the first pulse was probed and recorded from the second pulse after 440 nanoseconds. In addition to the lattice response, liquid tantalum scattering was measured for the first time under extreme conditions.

## Ongoing Innovation

Near the site where Lawrence’s four-meter synchrotron once rocketed atomic particles to 100 megaelectronvolts, the Advanced Light Source (ALS) at Lawrence Berkeley National Laboratory in Berkeley, California, now generates 1.9 gigaelectronvolts traveling at nearly the speed of light while emitting infrared, ultraviolet, and x rays through 40 beamlines. Since it first opened in 1993, Livermore scientists have used ALS for a range of in situ, time-resolved experiments. The 8.3.2 beamline at ALS uses x rays between 6 and 43 kiloelectronvolts (0.2 to 0.03 nanometers) and produces nondestructive 3D imaging of solid objects at a resolution down to 1 micrometer via computed tomography—the process of capturing a series of layered digital images of an object. Combining ALS’s synchrotron radiation microtomography and in situ uniaxial compression testing, Livermore scientists Holly Carlton, Nickolai Volkoff-Shoemaker, Mark Messner (now at ANL), Nathan Barton, Jon Lind, and Kumar have recently uncovered local microstrain deformations and failure modes of additively manufactured metal-lattice structures including the octet-truss, the rhombic-dodecahedron, and isotruss with different densities. They also utilized in situ computed tomography to incorporate 3D-defect distributions of lattice structures into model predictions for more detailed understanding of how these structures deform. “In the last decade, additive manufacturing or 3D printing has made a real impact on how we approach component development for the life-extension programs. We can design and build these complex, additively manufactured structures, but we need to know how they will respond under a range of conditions. Using high-resolution computed tomography at the ALS, we can map out defects, see how materials and configurations deform in real time, and use those findings to inform our design process and computational modeling,” says Lind, a Livermore physicist who has contributed to both quasi-static and dynamic testing.

Just as Lawrence knew that his “atom smasher” would be eclipsed by future technological innovations, contemporary light-source facilities continue to pursue and refine the next generation of tools that will uncover the secrets of the atomic world around us. ALS is currently undergoing an upgrade to produce highly focused soft x rays that will be at least 100 times brighter than the current beams. In 2024, APS will undergo a major, three-year upgrade to improve its capabilities, helping to keep the United States at the forefront of hard x-ray science. When completed, APS will produce the world’s brightest hard x rays and allow researchers to observe individual atoms moving and interacting in real time. LCLS at Stanford University is also preparing for a major upgrade—LCLS-II—which will increase its x-ray pulse repetition rate from 120 pulses per second to 1 million pulses per second and revolutionize how scientists study rare chemical events, quantum materials, and biological systems.

“We explore the behavior of materials at never-before-seen conditions, and Livermore teams have, in several instances, been the first to observe interesting behaviors, even in materials once thought to be well studied or boring,” says Gorman. “My colleagues and I are excited to know we might be able to synthesize novel materials or develop tools that could change the world.” Evans adds, “These x-ray light sources are an important, dramatically enhanced capability that are transformative for science around the world and for Livermore research.”

*—Genevieve Sexton*

**Key Words:** Advanced Light Source (ALS), Advanced Photon Source (APS), copper, cyclotron, Deutsches Elektronen-Synchroton (DESY), dynamic diamond anvil cell (dDAC), Dynamic Compression Sector (DCS), European XFEL (Eu-XFEL), gold, hard x rays, High-Pressure Collaborative Access Team (HPCAT), Linac Coherent Light Source (LCLS), nano diamond, nitroamine CL-20, Pohang Accelerator Laboratory X-ray Free-Electron Laser (PAL-XFEL), potassium, synchrotron, x-ray free electron laser (XFEL), zirconium.

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